#### Proceedings of the Fifth Lunar Conference (Supplement 5, Geochimica et Cosmochimica Acta) Vol. 2 pp. 2185–2203 (1974) Printed in the United States of America

## Solar flare and lunar surface process characterization at the Apollo 17 site\*

L. A. RANCITELLI, R. W. PERKINS, W. D. FELIX, and N. A. WOGMAN Battelle, Pacific Northwest Laboratories, Richland, Washington 99352

Abstract—The Apollo 17 lunar samples, of which 13 are considered here, have been analyzed for a wide spectrum of cosmogenic and primordial radionuclides. The intense solar flares of August 4–7, 1972 produced unusually high concentrations of the short-lived radionuclides; <sup>48</sup>V (16d), <sup>51</sup>Cr (28d), <sup>7</sup>Be (53d), <sup>56</sup>Co (78d), <sup>46</sup>Sc (84d), and substantially increased the concentration of <sup>54</sup>Mn (312d), <sup>22</sup>Na (2.60 yr) and <sup>60</sup>Co (5.24 yr). Based on very careful and accurate measurements of these flare-produced radionuclides, the intensity and energy spectrum of the August 1972 flare at the Apollo 17 site has been established. The time integrated proton flux above 10 MeV was  $7.9 \times 10^9$  protons/cm<sup>2</sup>. This flux was much harder than flares during the past five years with 36% of the proton flux above 30 MeV and 20% above 60 MeV. Expressed as a power function,  $E^{-\alpha}$ , the value of  $\alpha$  was 1.9. This is a substantially more rigid spectrum than the average for the past million years which is estimated to have an  $\alpha$  value of 3.1. Measurements of high- and low-energy reaction products in lunar samples exposed at different angles on the moon provide strong evidence for substantial angular anisotropy of the solar flare protons. The hardness of the solar flare proton flux increases dramatically as the incident angle moves from normal to low angles.

Soil which was permanently shadowed by a large boulder at Station 6 allowed the residence time of that boulder to be established at about one-half million years. The range in primordial radionuclide ratios was greater in samples at the Apollo 17 site than at any of the previous landing sites. Potassium to uranium ratios as high as 2860 and 6500 were observed for lunar soil and basalt, respectively, while many of the rock samples showed thorium to uranium ratios of less than 3.

#### Introduction

THE APOLLO 17 LUNAR MATERIAL provides a unique and precious set of samples not only because of the interesting geographical area which they represent, but also because of the spectrum of short-lived radionuclides which they contain. These radionuclides were produced by the two giant solar flares of August 4–7, 1972. This resulted in a very high proton flux on the lunar surface at the Apollo 17 site which was at that time 5 days into the lunar night. The relatively high concentrations of the short-lived cosmogenic radionuclides allowed their measurement with a high degree of accuracy which was sufficient to establish the energy spectrum of the incident proton flux and to provide an indication of its angular anisotropy.

Analyses of lunar samples from previous Apollo missions have already provided a great deal of information concerning the history and processes of the moon and sun (Perkins et al., 1970; Rancitelli et al., 1971, 1972; Finkel et al., 1971). The cosmogenic radionuclides reflect both the recent and long-term character of the solar and galactic cosmic ray flux, while primordial radionuclides help to

<sup>\*</sup>This paper is based on work supported by the National Aeronautics and Space Administration, Johnson Space Center, Houston, Texas under Contract NAS 9-11712.

describe the magmatic differentiation processes at the lunar surface. Apollo 11 sample studies provided our first observation of cosmogenic radionuclide production rates on the moon and confirmed the fact that the top centimeter of the lunar surface received a relatively intense periodic solar bombardment with a much smaller continuous galactic cosmic ray contribution (Perkins et al., 1970; Shedlovsky et al., 1970; O'Kelley et al., 1970). Measurements on core samples and other soil and lunar rock specimens from the Apollo 12 mission provided our first determination of erosion rate of lunar rocks, mixing of the lunar soil, together with an indication of the long-term average galactic cosmic ray flux and that of the recent solar flare (Rancitelli et al., 1971; Finkel et al., 1971). Apollo 15 samples provided several examples of relatively short exposure times, on the order of one-half million years and less. Samples from Apollo 16 were of a very unusual chemistry resulting in very different cosmogenic radionuclide ratios as a result of differences in target element abundance. A quiet sun for a long period preceding this mission allowed the measurement of ambient levels of galactic cosmic ray-produced short-lived radionuclides, while the friable soil clods collected by rake sampling established the fact that these materials with short lunar surface age—on the order of 10<sup>5</sup> yr—are evidently common.

Receipt of Apollo 17 samples for analysis at our laboratory began only 6 days after splashdown and this has permitted their measurement with the minimum of decay loss. The very high degree of accuracy and precision obtained in our earlier measurements (Perkins et al., 1970; Rancitelli et al., 1971, 1972) was maintained in these analyses and has allowed the observation of subtle differences in radionuclide concentrations. This high precision and accuracy has been exceedingly helpful as it has permitted small, but very meaningful variations of the cosmogenic radionuclides in soils and rocks to be observed.

### **PROCEDURE**

The multiple gamma coincidence counting techniques (Perkins et al., 1970) described in our earlier work were used to analyze the Apollo 17 samples. Briefly, the technique involves counting the sample for 8000–12,000 min in anticoincidence shielded multidimensional NaI(Tl) gamma-ray spectrometers. A comparison of the observed concentrations after background subtraction with the count rate of a sample mockup containing known radionuclide additions provides the basis for determining the radionuclide concentrations.

The mockups are prepared from a mixture of casting resin, iron powder, and aluminum oxide that contains a precisely known radionuclide addition. These mockups reproduce the precise shape, size, physical and electron densities of the samples. In most cases, the uncertainties associated with counting statistics were on the order 1–2% for <sup>22</sup>Na, <sup>26</sup>Al, K, Th, and U while the absolute errors for these radionuclides based on all analytical uncertainties including the error in radioisotope standards ranged from 3% to 8%. Precise mockups were not available for all the Apollo 17 samples analyzed. In these cases, the counting efficiencies and Compton corrections were estimated from our library of over 100 lunar mockups of various sizes and shapes by the methods used by O'Kelley *et al.* (1970). We have assigned appropriately larger uncertainties to these determinations. These values will be refined when sample models become available. Because of the relatively high concentrations of the short-lived radionuclides <sup>46</sup>Sc, <sup>54</sup>Mn, and <sup>56</sup>Co, it was possible to achieve similar accuracies. For the measurement of the short-lived cosmogenic radionuclides, a large Ge(Li) diode with an annular sodium-iodide anticoincidence shield (Cooper and Perkins, 1972) proved very useful. Spectra of lunar rocks 75055

recorded with this analyzer are shown in Fig. 1. Two spectra are actually recorded simultaneously. The one, called a coincidence spectrum, is recorded where events are seen in the sodium-iodide crystal at the same time as an event in the Ge(Li) detector. This, of course, requires the simultaneous emission of two or more gammas. The anticoincidence spectrum is accumulated where only a single gamma ray is emitted by the radionuclide. The value of this technique for measuring short-lived radionuclides such as <sup>51</sup>Cr, <sup>7</sup>Be and <sup>54</sup>Mn which emit only one gamma ray is illustrated in Fig. 2 for rock 75055. The upper spectra show the results which would be achieved with a normal Ge(Li) diode, while the two lower sets of spectra are amplified portions of the anticoincidence spectra shown in Fig. 1. The improvement in ability to measure all of these radionuclides is obvious, and in the case of <sup>7</sup>Be and <sup>51</sup>Cr, their measurement without the anticoincidence technique would not be nearly as reliable.

#### RESULTS AND DISCUSSION

### Cosmogenic radionuclide concentrations

The cosmogenic radionuclide concentrations observed in the Apollo 17 lunar samples are summarized in Table 1. In Table 2 the concentrations of the cosmogenic radionuclides in a soil sample from each of the Apollo missions is compared. Excluding the analysis from Apollo 16, which had a very unusual chemistry, it is evident that the cosmogenic radionuclide concentrations of the Apollo 17 samples are strikingly different. One of the most obvious differences is the <sup>26</sup>Al to <sup>22</sup>Na ratio which was relatively constant at approximately two for the first four missions, yet is approximately one for the Apollo 17 samples. The target elements from which these radionuclides are produced have approximately the same concentrations in Apollo 17 soils as that in Apollo 11 and 15. It is thus evident that the solar flare of August 1972 produced approximately as much additional <sup>22</sup>Na in the lunar surface as was present just prior to the flare. The relatively high concentrations of <sup>46</sup>Sc result from a proton reaction on titanium, which itself was at a very high concentration in Apollo 17 soil. The cosmogenic radionuclides 54Mn, 56Co, 57Co, and 58Co result almost entirely from proton and alpha-particle spallation reactions on iron. Among the Apollo 17 samples (see Table 3) there is approximately a factor of 2 variation in both the iron and aluminum concentrations; therefore, an interpretation of the <sup>26</sup>Al content relative to the cosmogenic radionuclides resulting from spallation reactions on iron must be based on chemical composition. Since the radionuclides <sup>56</sup>Co and <sup>54</sup>Mn are produced almost entirely by spallation reactions on iron, the ratios of these radionuclides in lunar samples are essentially independent of chemical composition of the specimen. Because of their relatively high concentrations and the associated ease of accurate measurements, they serve here as important indicators in interpreting the characteristics of the solar flare.

### Lunar surface processes

Of the 23 Apollo 17 lunar samples which have been received at our laboratory at this writing, the analyses of 13 are sufficiently well analyzed to include in this discussion. These include 3 samples from Station 1; 2 from Station 5; 5 from

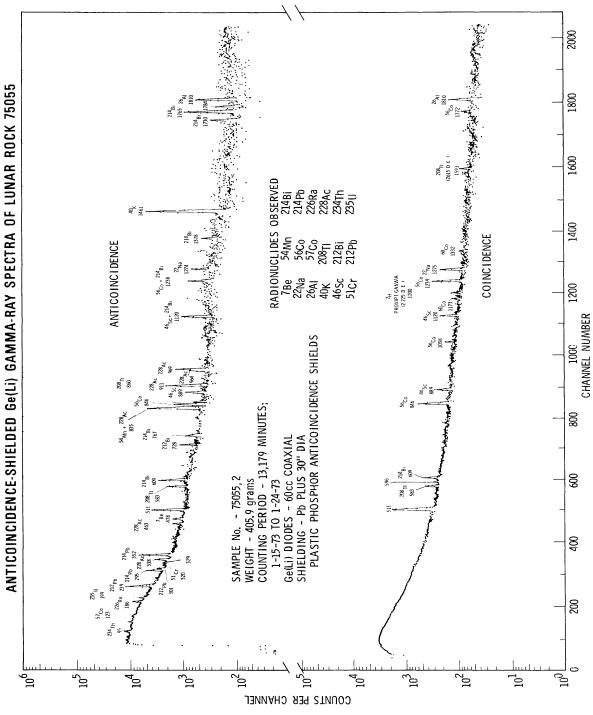


Fig. 1. Anticoincidence shielded Ge(Li) gamma-ray spectra of lunar rock 75055.

### <sup>7</sup>Be, <sup>51</sup>Cr, AND <sup>54</sup>Mn REGIONS OF ANTI-COINCIDENCE SHIELDED Ge (Li) GAMMA RAY SPECTRA OF APOLLO 17 ROCK, 75055, 1

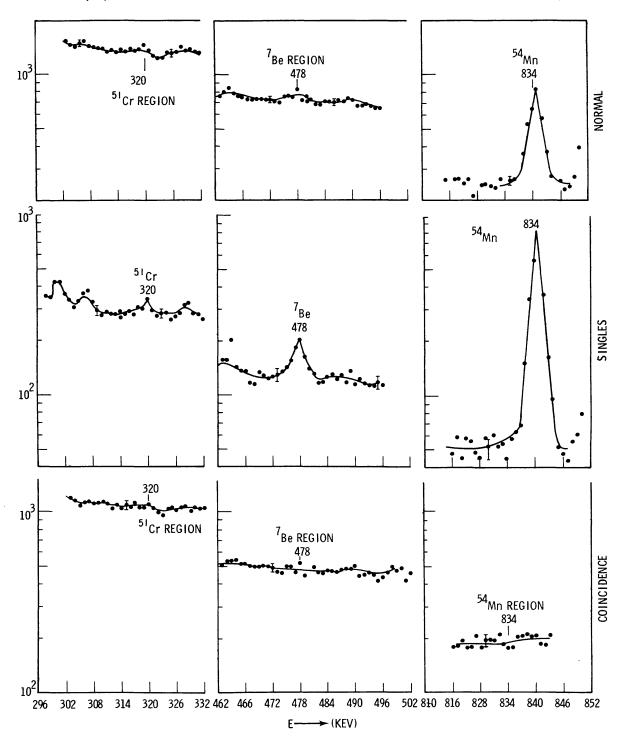


Fig. 2. <sup>7</sup>Be, <sup>51</sup>Cr, and <sup>54</sup>Mn regions of anticoincidence shielded Ge(Li) gamma-ray spectra of Apollo 17 Rock, 75055,1.

Table 1. Cosmogenic\* and primordial radionuclides in Apollo 17 materials.

Radionuclides	71035,0	71041,4	71155,0	75055,2	75061,5	76240,2
<sup>7</sup> Be (dpm/kg)		_		$140 \pm 20$	$350 \pm 120$	
<sup>22</sup> Na (dpm/kg)	$92 \pm 4$	$123 \pm 4$	$112 \pm 4$	$85 \pm 5$	$171 \pm 5$	$42\pm2$
<sup>26</sup> Al (dpm/kg)	$79 \pm 3$	$123 \pm 4$	$105 \pm 4$	$69 \pm 7$	$174 \pm 5$	$151 \pm 6$
46Sc (dpm/kg)	$87 \pm 5$	$75 \pm 3$	$80 \pm 4$	$62 \pm 7$	$112 \pm 5$	$8 \pm 4$
48V (dpm/kg)		$27 \pm 15$	<60	<22	$26 \pm 13$	2.6
51Cr (dpm/kg)				$75 \pm 37$	_	< 200
54Mn (dpm/kg)	$164 \pm 15$	$198 \pm 10$	$227 \pm 30$	$139 \pm 15$	$286 \pm 12$	$21 \pm 8$
<sup>56</sup> Co (dpm/kg)	$279 \pm 14$	$379 \pm 10$	$310 \pm 20$	$210 \pm 15$	$548 \pm 30$	$27 \pm 3$
<sup>57</sup> Co (dpm/kg)				$7.4 \pm 1.5$	$18\pm7$	<12
58Co (dpm/kg)				$6.9 \pm 3$	$21 \pm 14$	<12
60Co (dpm/kg)	<4.6	<2.2	<4.4	<3	<2.8	$0.8 \pm 0.4$
K (ppm)	$200 \pm 20$	$630 \pm 40$	<450	$650 \pm 50$	$600 \pm 30$	$1100 \pm 50$
Th (ppm)	$0.36 \pm 0.03$	$0.863 \pm 0.080$	$0.29 \pm 0.05$	$0.40 \pm 0.02$	$0.87 \pm 0.03$	$2.30 \pm 0.11$
U (ppm)	$0.096 \pm 0.011$	$0.25 \pm 0.01$	$0.13 \pm 0.02$	$0.10 \pm 0.01$	$0.22 \pm 0.01$	$0.60 \pm 0.03$
K/U	2450	2520	<3460	6500	2730	1830
Th/U	4.00	3.93	2.23	4.00	3.95	3.83
Sample weight						
(grams)	144.11	111.1	25.8	405.9	100.0	104.98
Sample type†	VFCB	Soil	VFGB	Vesicular	Soil, top	Soil,
				Basalt	of	permanent
					Boulder	Shadow

<sup>\*</sup>Decay corrected to December 17, 1972.

Radionuclides	76255,0	76261,1	76275,0	76295,0	77135,0	78421,1	78481,1
<sup>7</sup> Be (dpm/kg)					-	<u></u>	$370 \pm 90$
<sup>22</sup> Na (dpm/kg)	$71 \pm 4$	$143 \pm 4$	$100 \pm 3$	$64 \pm 3$	$100 \pm 5$	$39 \pm 2$	$244 \pm 12$
<sup>26</sup> Al (dpm/kg)	$79 \pm 4$	$171 \pm 5$	$110 \pm 3$	$71 \pm 4$	$111 \pm 6$	$55 \pm 2$	$257 \pm 12$
46Sc (dpm/kg)	$3.9 \pm 1.2$	$27 \pm 3$	$7\pm2$	$6.4 \pm 2.6$	$7.2 \pm 2.2$	$9.3 \pm 2.5$	$59 \pm 3$
48V (dpm/kg)	<4	<29			<4		$34 \pm 15$
<sup>51</sup> Cr (dpm/kg)		_	_	_			<340
54Mn (dpm/kg)	$38 \pm 9$	$106 \pm 14$	$103 \pm 20$	$69 \pm 26$	$21 \pm 15$	<42	$264 \pm 20$
<sup>56</sup> Co (dpm/kg)	$37 \pm 4$	$246 \pm 8$	$86 \pm 9$	$35 \pm 5$	$66 \pm 4$	<6	$606 \pm 30$
<sup>57</sup> Co (dpm/kg)	_		_			_	$18\pm3$
<sup>58</sup> Co (dpm/kg)		_		_			$18 \pm 12$
60Co (dpm/kg)	$2.5 \pm 0.5$	< 2.5	<1.1	<1.1	$3.4 \pm 1.5$	$1.6 \pm 0.8$	
K (ppm)	$2900 \pm 140$	$970 \pm 40$	$2250 \pm 90$	$2300 \pm 80$	$1850 \pm 90$	$840 \pm 30$	$950 \pm 40$
Th (ppm)	$2.33 \pm 0.10$	$1.92 \pm 0.04$	$5.69 \pm 11$	$5.76 \pm 0.17$	$5.51 \pm 0.28$	$1.58 \pm 0.07$	$1.49 \pm 0.0$
U (ppm)	$0.58 \pm 0.02$	$0.52 \pm 0.02$	$1.40 \pm 0.03$	$1.55 \pm 0.05$	$1.42 \pm 0.07$	$0.41 \pm 0.02$	$0.39 \pm 0.0$
K/U	5000	1865	1610	1480	1300	2050	2440
Th/U	4.01	3.69	4.06	3.72	3.88	3.86	3.82
Sample weight		3.02			2.00	- 700	2.02
(grams)	393.2	100.7	55.93	260.7	316.7	94.51	101.27
Sample type†	Breccia	Soil,	PRB	PRB	VHB	Soil,	Soil,
		2 cm skim				15–25 cm	1 cm ski

<sup>†</sup>VFGB-Very fine-grained basalt.

PRB—Partially recrystallized breccia.

VHB-Vesicular hemfelsic breccia.

	(dpm/kg)							
	10084,41	12070,13	14163,0	15041,14	69941,1	71041,4		
<sup>22</sup> Na	64 ± 3	$80 \pm 2$	$44 \pm 2$	$65 \pm 2$	$39 \pm 2$	123 ± 4		
<sup>26</sup> Al	$131 \pm 4$	$165 \pm 4$	$89 \pm 4$	$123 \pm 4$	$147 \pm 5$	$123 \pm 4$		
<sup>26</sup> Al/ <sup>22</sup> Na	$2.05 \pm 0.11$	$2.06 \pm 0.07$	$2.02 \pm 0.13$	$1.89 \pm 0.08$	$3.77 \pm 0.23$	$1.00 \pm 0.04$		
<sup>46</sup> Sc	$11 \pm 1$	$5.9 \pm 1.6$	< 5.3	$2.9 \pm 1.6$	< 6.9	$75 \pm 3$		
$^{48}V$	<10			<10		$27 \pm 15$		
<sup>54</sup> Mn	$24 \pm 3$	$21 \pm 8$	<38	$14 \pm 8$	$37 \pm 14$	$198 \pm 10$		
<sup>56</sup> Co	$53 \pm 10$	$57 \pm 7$	<13	$27 \pm 6$	<23	$379 \pm 10$		

Table 2. Cosmogenic radionuclide content of Apollo soils.

Station 6; 1 from Station 7; and 2 from Station 8, consisting of 3 basalt samples, 4 breccia, and 6 soils. The <sup>26</sup>Al to <sup>22</sup>Na ratio for most of the surface samples is approximately 1, and significant deviations from this ratio may suggest a short exposure age relative to the half-life of <sup>26</sup>Al (0.74 m.y.), or partial shielding relative to  $2\pi$  irradiation geometry. Three lunar samples were analyzed from Station 1. They include chips (71035,0 and 71155,0) from the top surface of boulders approximately one meter apart and one soil sample (71041,4). The <sup>26</sup>Al to <sup>22</sup>Na ratios in each of these were essentially 1, indicating a lunar surface exposure long compared with the half-life of <sup>26</sup>Al. Samples from Station 6 present a very interesting picture, since they indicate the boulders in this area have attained their present position within the past million years. Photographic documentation of this area clearly showed tracks of 3 large boulders which they had left in rolling

Table 3. Elemental composition of Apollo 17 specimens.\*

Sample No.	Si	Al	Ca	Mg	Ti	Fe	Na	Mn	K	S	P	Oª
71041,4	18.58	5.72	7.66	5.86	5.74	13.78	0.26	0.19	0.07	0.13	0.03	41.98
71155,0 <sup>b</sup>	17.38	4.59	7.45	5.14	7.88	15.25	0.24	0.22	0.03	0.18	0.04	41.60
75055,2	19.29	5.16	8.79	4.13	6.10	14.18	0.33	0.22	0.07	0.19	0.03	41.51
75061,5	18.38	5.51	7.66	5.75	6.18	14.14	0.24	0.19	0.07	0.13	0.03	41.72
76215,0°	21.56	9.53	7.88	7.62	0.92	7.08	0.39	0.10	0.25	0.80	0.12	43.75
76240,2°	20.29	9.86	8.78	6.68	1.89	8.02	0.26	0.11	0.08	0.07	0.03	43.93
76261,1°	20.29	9.86	8.78	6.68	1.89	8.02	0.26	0.11	0.08	0.07	0.03	43.93
76255,0 <sup>d</sup>	21.42	9.53	7.90	7.48	0.88	6.95	0.42	0.09	0.22	0.08	0.13	44.90
$76275,0^{d}$	21.42	9.53	7.90	7.48	0.88	6.95	0.42	0.09	0.22	0.08	0.13	44.90
$76295,0^{d}$	21.42	9.53	7.90	7.48	0.88	6.95	0.42	0.09	0.22	0.08	0.13	44.90
77135,0	21.56	9.53	7.88	7.62	0.92	7.08	0.39	0.10	0.25	0.80	0.12	43.75
78481 <sup>t</sup>	19.95	8.33	8.41	5.98	3.28	10.22	0.26	0.14	0.07	0.10	0.02	43.24

<sup>\*</sup>Lunar Sample Information Catalog, Apollo 17 (1973).

<sup>\*</sup>By difference.

<sup>&</sup>lt;sup>b</sup>Composition of sample 70215 assumed. See text.

<sup>&</sup>lt;sup>c</sup>Composition of sample 77135 assumed. See text.

<sup>&</sup>lt;sup>d</sup>Composition of sample 76315 assumed. See text.

<sup>\*</sup>Composition of sample 76501,2 assumed. See text.

<sup>&</sup>lt;sup>t</sup>Composition of sample 78501,2 assumed. See text.

down an adjacent hillside. The lunar soil samples 76261,2 and 76240,2 both indicate shielding and undersaturation with respect to  $^{26}$ Al. Soil sample 76240,2 was collected beneath an overhang on a very large boulder in a permanently shielded area, while sample 76261,1 was collected about one meter from the boulder. Based on the target element concentrations of 76261,1 a saturation concentration for  $^{26}$ Al would be approximately 250 dpm/kg. The much lower observed concentration of 171 dpm/kg is attributed to a 60% shielding relative to the  $2\pi$  irradiation that it would have been exposed to if it were on a flat plain for the past several million years. The highly shadowed lunar sample, 76240, is shielded from about 85% of the  $2\pi$  radiation it would have been exposed to were it on a flat plain, as indicated by the  $^{22}$ Na concentration of only 42 dpm/kg. A similar analysis of the  $^{26}$ Al excess for its present shielding indicates that it has been shielded by the adjacent boulder for a period of approximately one-half million years. This interpretation has also been suggested by Keith *et al.* (1973) for the high  $^{26}$ Al to  $^{22}$ Na ratio for soil sample 76240.

Sample 78421,1 is a trench soil sample from Station 8. Its depth, which is greater than 15 cm, shielded it almost completely from the August 1972 solar flare and the concentrations are therefore what one would predict from the galactic cosmic ray component (Rancitelli *et al.*, 1971).

The cosmogenic radionuclides <sup>56</sup>Co and <sup>54</sup>Mn are spallation products of iron and their concentrations are thus dependent on chemical composition as well as the geometry of the sample on the lunar surface. As we will consider in the following discussion, the ratios of these radionuclides can provide direct information on the rigidity of the solar flare and the angular anisotropy; however, since the radionuclide ratio is also a function of the angle of the exposure surface of lunar samples, it can provide direct information on the sample geometry as well.

In our previous work (Rancitelli et al., 1971), we described a model for calculating the depth dependence of <sup>26</sup>Al and <sup>22</sup>Na production from a representative incident solar proton spectrum, the appropriate excitation functions and the sample's target element composition. This model employs the kinetic power law form:

$$\frac{dJ}{dE} = kE^{-\alpha} \tag{1}$$

to describe the incident proton spectrum, where J is the proton flux, E is the proton energy,  $\alpha$  is a shape function, and k a constant related to the particle intensity. Using this model, we have calculated the best fit for the observed <sup>26</sup>Al gradients in solar and rock samples to be 3.1. The concentration gradient for <sup>22</sup>Na in Apollo 12 core samples and rock sections also showed an alpha equal to 3.1. Thus, it appeared that the solar flares which had produced <sup>22</sup>Na in lunar material had the same energy shape function as the average for the past few million years. An excellent pair of radionuclides for determining the rigidity of the August 1972 solar flare is <sup>56</sup>Co and <sup>54</sup>Mn. Both of these are produced almost entirely from iron, yet <sup>56</sup>Co is a very low-energy product, whereas <sup>54</sup>Mn is a relatively high-energy product with a threshold of 25 MeV (see Fig. 3). Other radionuclides, including



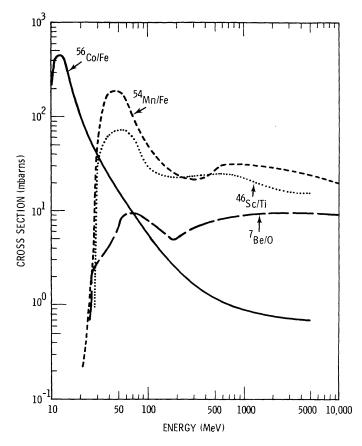


Fig. 3. Excitation functions for production of <sup>7</sup>Be, <sup>46</sup>Sc, <sup>54</sup>Mn, <sup>56</sup>Co from major target elements.

<sup>46</sup>Sc and <sup>7</sup>Be, can serve to verify the energy spectrum estimate based on the <sup>56</sup>Co to <sup>54</sup>Mn ratio. In Fig. 4 the calculated ratios of <sup>56</sup>Co to <sup>54</sup>Mn are plotted as a function of sample thickness for alpha values ranging from 1.2 to 4. Also we have indicated our observed values for 4 Apollo 17 lunar soil samples, for rock sample, 75055, and for the Apollo 11 soil sample 10084. For this analysis the contribution from galactic cosmic rays to <sup>54</sup>Mn and <sup>56</sup>Co production was subtracted using galactic cosmic ray production rates from our earlier work (Perkins *et al.*, 1970). From the soil samples, we have established the best fit for alpha as 1.9 for the August 1972 solar flare. Thus, the proton energy spectrum for the August solar flare was much more rigid than that of both previous recent flares and of the average for the past million years. The average rigidity of the flares which preceded the Apollo 11 mission was about 3.2 as indicated by plotting our value for the Apollo 11 soil 10089 (Perkins *et al.*, 1970) on Fig. 4.

In Figs. 5, 6, and 7 the expected concentrations of <sup>7</sup>Be, <sup>22</sup>Na, <sup>46</sup>Sc, <sup>51</sup>Cr, <sup>54</sup>Mn, and <sup>56</sup>Co have been calculated as a function of depth for an alpha value of 1.9 for the lunar soil samples 75061 and 78481 and rock chip sample 75055. We have also plotted the observed concentrations of these radionuclides on the curves. The

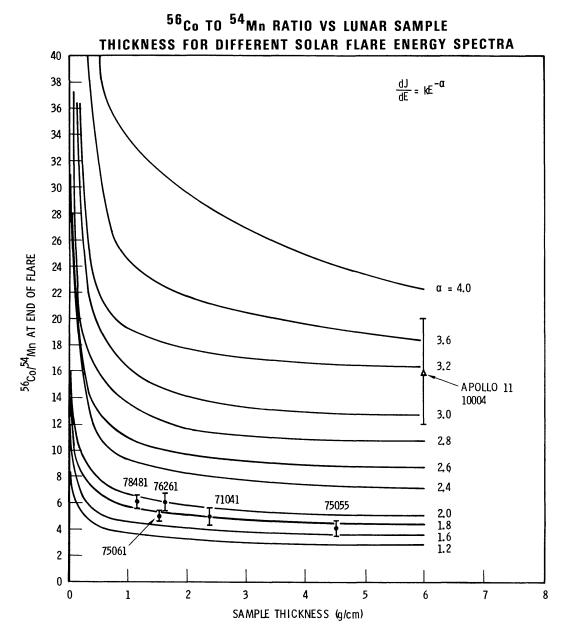


Fig. 4. <sup>56</sup>Co to <sup>54</sup>Mn ratio versus lunar sample thickness for different solar flare energy spectra.

observed radionuclide concentrations in the two soil samples fit the calculated data very well, indicating good agreement with the alpha value of 1.9. For rock sample 75055, the concentrations were all low by approximately 60%. For the comparison, we have therefore normalized the curves so that the observed <sup>46</sup>Sc, <sup>54</sup>Mn, <sup>56</sup>Co, and <sup>22</sup>Na best fit to the calculated lines. The relatively low concentrations of radionuclides in this rock sample are apparently due to the fact the sample was taken from the southeast face of the boulder which was inclined about 45° to the horizon, and thus received a substantially lower integrated exposure than the relatively flat lunar soil samples.

From Eq. (1) and the radionuclide concentrations in the lunar soil, it is possible

### COMPARISON OF CALCULATED TO OBSERVED RADIONUCLIDE CONTENT OF APOLLO 17 SOIL 75061

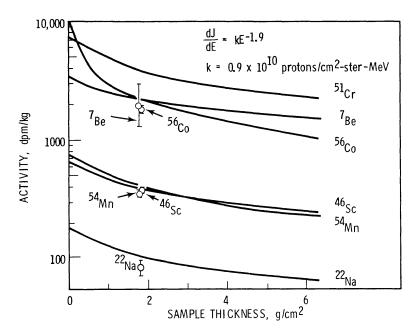


Fig. 5. Comparison of calculated to observed radionuclide content of Apollo 17 soil 75061.

### COMPARISON OF CALCULATED TO OBSERVED RADIONUCLIDE CONTENT OF APOLLO 17 SOIL 78481

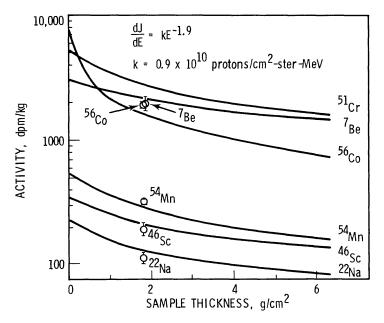


Fig. 6. Comparison of calculated to observed radionuclide content of Apollo 17 soil 78481.

### COMPARISON OF CALCULATED TO OBSERVED RADIONUCLIDE CONTENT OF APOLLO 17 ROCK 75055

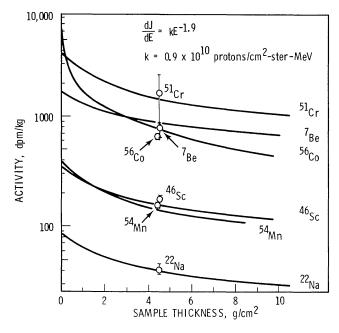


Fig. 7. Comparison of calculated to observed radionuclide content of Apollo 17 rock 75055.

to calculate the total proton flux from the August flare. This flux and the associated energy shape function are included in Fig. 8. Included in Fig. 9 are similar data by King (1973) back through 1965. As indicated in the figures, the integral proton flux above 10 MeV from the August solar flare of 7.8 × 10° protons/cm² far exceeds the intensity and rigidity of the recent solar flares. This can be compared to the value of 1.4 × 10° protons from the January 1971 flare (Rancitelli et al., 1971). Integration of energy areas about 30 and 60 MeV, respectively, show 36% and 20% of the protons in these areas. In Table 4 we have compared our integrated proton fluxes with those reported by King (1972) and Bostrum et al. (1972) from satellite measurements. Therefore, in Table 5 we have summarized all of our observed <sup>56</sup>Co to <sup>54</sup>Mn ratios for the Apollo 17 samples and the approximate angle of the surface of the rock samples relative to the horizontal are presented. While the surface angle of these samples is only an estimate from

Table 4. Integrated proton flux for the August 4–7, 1972 solar flares.  $J: (10^8 \text{ protons/cm}^2\text{-ster.})$ 

Energy interval (MeV)	This work	King (1972)	Bostrum <i>et al.</i> (1972)
>10	12.6	11	16
>30	4.6	6.1	6.4
>60	2.5	2.0	1.9

### COMPARISON OF TIME INTEGRATED FLUX OF AUGUST 4-7, 1972 SOLAR FLARE WITH PREVIOUSLY REPORTED SOLAR FLARES

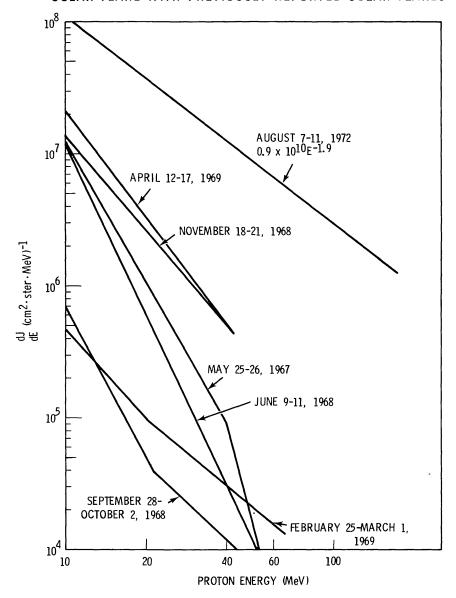


Fig. 8. Comparison of time integrated flux of August 4–7, 1972 solar flare with previously reported solar flares (King 1973).

the lunar sample catalog, it is sufficient to indicate the extreme dependence of the ratio of these isotopes on the surface sample's orientation.

From our analysis of the proton energy spectrum (Fig. 4) we selected mainly samples which had been exposed on a flat plain (i.e. 4 soils). Here we observed <sup>56</sup>Co to <sup>54</sup>Mn ratios of 5 to 6. Examination of the <sup>56</sup>Co to <sup>54</sup>Mn ratios in other samples indicated that information on angular anisotropy could also be obtained. The samples that had their surface at the greatest angle relative to the horizontal plain showed the lowest <sup>56</sup>Co to <sup>54</sup>Mn ratio. This ratio is, of course, very sensitive to the hardness of the incident solar flare energy spectrum (see Fig. 4). For

### **MAJOR SOLAR FLARE INTENSITIES**

 $J_i > 10 \text{ MeV}$ 

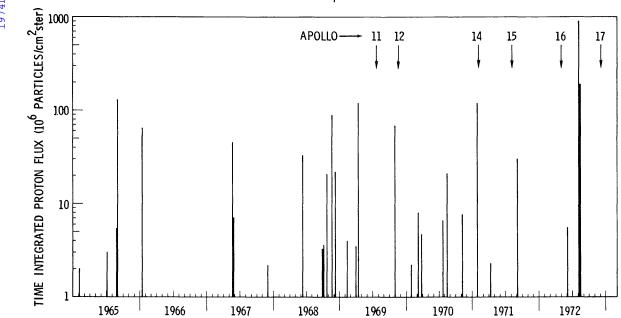


Fig. 9. Major solar flare intensities from 1965 to 1972 (King, 1972).

example, the 3 boulder chips 76255, 76275, and 76295, which had high surface angles relative to the horizon, had very low <sup>56</sup>Co to <sup>54</sup>Mn ratios. The implication of these observations is that there is a much lower ratio of low- to high-energy protons arriving at the lunar surface at low angles. Thus, a high degree of anisotropy exists which is a function of the incident angle. Sample 76295,0 was taken from an essentially vertical surface and had one of the lowest observed <sup>56</sup>Co to <sup>54</sup>Mn ratios, 1.85. This would be comparable to an alpha value of less than 1 (see Fig. 4) thus an extremely rigid spectrum. Anisotropy is also indicated in soil sample 76240 taken from beneath the overhang of the very large boulder at Station 6. Here, the <sup>56</sup>Co to <sup>54</sup>Mn ratio of 2.54 is much lower than the value of 6 in the adjacent solar sample 76261,1. Since only protons of low incident angle could strike soil sample 76240,2, this is substantiating evidence of anisotropy favoring high-energy protons at low angles. While all of the boulder chip samples were taken from southeast or east faces of lunar surface boulders, soil sample 76240 was shielded except from the north. Thus, angular anisotropy appears to be independent of compass direction.

The energy spectrum which impinged at different angles of lunar surface orientation can be quantified by integrating the solar proton flux over several energy intervals and employing the proton spectrum hardness factor alpha from Eq. (1) for that angle. When this is done over the energy interval 30–100 MeV and compared with the total number of protons present in the flare (between 10 and 100 MeV), this defines the relative number of protons with energies higher than the <sup>54</sup>Mn production threshold. Results of these calculations are summarized in Table 6 for lunar surface orientation between 0° and 90°. The alpha value

Table 5. Relationship of solar cosmic ray-induced <sup>56</sup>Co and <sup>54</sup>Mn to lunar surface orientation.\*

Sample No. (Type)	<sup>56</sup> Co (dpm/kg)	<sup>54</sup> Mn (dpm/kg)	<sup>56</sup> Co/ <sup>54</sup> Mn	Lunar surface† orientation
71035,0 (Boulder chip)	$870 \pm 40$	$190 \pm 20$	4.58	SE, 15°
71041,4 (Soil)	$1130 \pm 30$	$220 \pm 13$	5.13	0°
71155,0 (Boulder chip)	$960 \pm 60$	$240 \pm 40$	4.00	SE, 30°
75055,2 (Boulder chip)	$650 \pm 47$	$160 \pm 16$	4.06	SE, 45°
75061,5 (Soil)	$1700 \pm 90$	$340 \pm 16$	5.00	0°
76240,2 (Soil)	$84 \pm 9$	$33 \pm 7$	2.55	N, 0°
76255,0 (Boulder chip)	$120 \pm 12$	$38 \pm 12$	3.16	SE, 60°
76261,1 (Soil)	$760 \pm 25$	$130 \pm 19$	5.97	0°
76275,0 (Boulder chip)	$270 \pm 19$	$120 \pm 26$	2.25	SE, 60°
76295,0 (Boulder chip)	$120 \pm 16$	$62 \pm 29$	1.94	SE, 90°
78135,0 (Rock)	$900 \pm 90$	$180\pm20$	5.00	0°
78481,1 (Soil)	$1880 \pm 90$	$310 \pm 13$	6.06	0°

<sup>\*</sup>Decay corrected to August 7, 1972.

Table 6. Relationship of lunar sample surface orientation at the Apollo 17 site to the proton energy spectrum of the August 1972 solar flare.

Lunar surface orientation (degrees)*	α	$\frac{J_i (30 > Ep > 100 \text{ MeV})}{J_i (10 > Ep > 100 \text{ MeV})}$
0°	$2.0 \pm 0.1$	$0.26 \pm 0.02$
15°	$1.8\pm0.2$	0.29
30°	$1.7\pm0.1$	0.33
45°	$1.7 \pm 0.2$	0.31
60°	$1.4 \pm 0.2$	0.41
90°	$0.9 \pm 0.3$	0.55
0° (Apollo 11)	$3.2 \pm 0.4$	$0.08 \pm 0.04$
0° (m.y. Average)	$3.0\pm0.2$	$0.10 \pm 0.02$

<sup>\*</sup>Angle to the horizontal.

<sup>†</sup>Lunar compass direction of sample surface and angle of surface to horizontal.

decreases from 2.0 for sample surfaces at 0° to 0.9 for sample surfaces at 90°. The fraction with energies above 30 MeV is approximately 26% at 0° and increases to 55% for orientations of 90°. As an indication of the extreme hardness of August 1972 flares these proton ratios are compared with those calculated for the April 1969 flare which preceded the Apollo 11 landing. This 1969 flare had an average alpha of 3.2 with only 8% of the proton having energies in excess of 30 MeV. These alpha values can be compared with the long-term average value of 3.0 which we previously determined from <sup>26</sup>Al gradients in soil and rock samples. Thus for the last one or two million years, 10% of the protons have energies of greater than 30 MeV.

#### Primordial radionuclides

The primordial radionuclide content of lunar samples provides basic information on the moon's geochemistry, on local transport and mixing of soil, and on the moon's relationship to other bodies in the solar system. Where sufficient and properly selected samples are studied, their primordial radionuclide content may also serve to define stratigraphic relationships through core sample analysis (Rancitelli *et al.*, 1970) and also the relationship of breccia and crystalline rocks to their surrounding and underlying soil.

Analysis of samples from the Apollo 17 site show far greater differentiation of primordial radionuclide ratios than has been observed in samples from any of the other previous missions. The primoridal radionuclide concentrations and their ratios are summarized in Table 1 along with the cosmogenic radionuclides. For comparison, the potassium to uranium content of the Apollo 17 samples analyzed at our laboratory are plotted in Fig. 10 along with most of the large sample analyses from previous missions (Perkins et al., 1970; O'Kelley et al., 1970, 1971; Rancitelli et al., 1971, 1972). Table 7 compares the primordial radionuclide concentrations in lunar soil samples for all of the Apollo missions. From the comparisons in Table 7, it is evident that higher potassium to uranium ratios were observed in soil samples from the Apollo 17 site than at any of the other sites. These soil samples were taken from Stations 1 and 5 where potassium to uranium ratios of 2860 and 2730, respectively, were observed. One of the more spectacular observations was the very high potassium to uranium ratio of 6500 in the basalt

Table 7. Comparison of primordial radionuclide concentrations in lunar soils from the Apollo mission.

	K (ppm)	Th (ppm)	U (ppm)	K/U
Apollo 11	1100	2.3	0.55	2000
Apollo 12	2000	6.7	1.7	1180
Apollo 14	4400	14.6	3.6	1220
Apollo 15	1200-1700	3.8-4.7	0.94-1.3	1200-1500
Apollo 16	740-1200	1.8-2.7	0.41 - 0.74	1470-2070
Apollo 17	600-1100	0.86-2.3	0.22 - 0.60	1830-2860

# POTASSIUM vs URANIUM CONTENT OF LUNAR MATERIALS

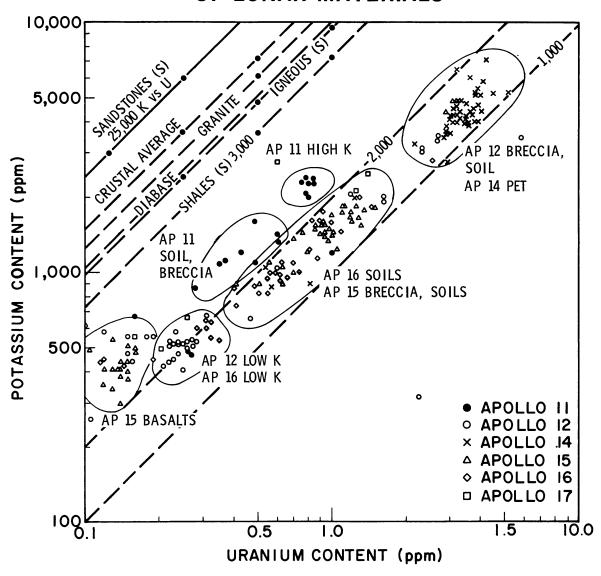


Fig. 10. Comparison of K versus U content of Apollo 17 samples with previous Apollo samples and terrestrial materials.

sample 75055. This is considerably higher than has been observed in any of the previous Apollo samples, thus substantiating an unusual differentiation process at this site. Another indication of major differentiation is evidenced by the very low thorium to uranium ratios in several of the Apollo 17 rock samples. The lowest observed ratio is 2.3 for the basalt rock chip 71155; however, values of three and less were observed in six other rock samples studied by the preliminary examination team (Lunar Sample Information Catalog, Apollo 17). Lunar rock samples from previous Apollo missions had shown an atomic ratio for <sup>232</sup>Th: <sup>238</sup>U of about 3.8, which is the average for our present-day solar system (Fowler and Hoyle, 1960). The average thorium to uranium atomic ratio for the Apollo 17 soils

was 3.95. As has been stated earlier (Perkins et al., 1970), the soil values are in some question. This is due to the fact that radon loss could produce a negative bias in the uranium value, thus giving an artificially high thorium to uranium ratio. We believe that this bias is less in the more recent Apollo missions because of the Curator's practice of sealing the samples in double teflon bags which should help minimize radon loss. The lowest value of potassium observed in the Apollo 17 samples was 200 ppm in the basalt chip 71035. Except for a few samples from the Apollo 16 mission, and rock 12005 from the Apollo 12 mission, this is the lowest observed value for a lunar sample. From the sites where we have thus far analyzed both soil and rock samples, there was a correlation between their relative primordial radionuclide contents. For example, Station 1 showed low concentrations in both the soil and rock samples, while Station 6 showed much higher concentrations in both soil and rock samples. Station 5 showed intermediate values for the soil and rocks.

However, in all cases the primordial radionuclide ratios are substantially different in the rocks than in the surrounding soils, thus making the relationship of the soil to the rocks a difficult problem.

Acknowledgments—We wish to thank R. M. Campbell, D. R. Edwards, J. G. Pratt, and J. H. Reeves of this Laboratory for their aid in standards preparation and in data acquisition. The unique and sensitive instrumentation that made this work possible was developed during the past decade under sponsorship of the United States Atomic Energy Commission, Division of Biomedical and Environmental Research.

#### REFERENCES

- Bostrom C. O., Kohl J. W., McEntire R. W., and Williams D. J. (1972) The solar proton flux—August 2-12. Preprint—Applied Physics Laboratory (November 1972).
- Cooper J. A. and Perkins R. W. (1972) A versatile Ge(Li)-NaI(Tl) coincidence—anticoincidence gamma-ray spectrometer for environmental and biological problems. *Nucl. Instr. & Methods* 99(1), 125.
- Finkel R. C., Arnold J. R., Imamura M., Reedy R. C., Fruchter J. S., Loosli H. H., Evans J. C., Delany A. C., and Shedlovsky J. P. (1971) Depth variation of cosmogenic nuclides in a lunar surface rock and lunar soil. *Proc. Second Lunar Sci. Conf.*, *Geochim. Cosmochim. Acta*, Suppl. 2, Vol. 2, pp. 1773–1789. MIT Press.
- Fowler W. A. and Hoyle F. (1960) Nuclear cosmochronology. Ann. Phys. 10, 280.
- Keith J. E. and Clark R. S. (1973) Personal communication.
- King J. H. (1972) Personal communication (September 29).
- King J. H. (1972) Study of mutual consistency of IMP 4 solar proton data. NSSDC 72-14 (October).
- Lunar Sample Information Catalog, Apollo 17 (1973). National Aeronautics and Space Administration MSC 03211.
- O'Kelley G. D., Eldridge J. S., Schonfeld E., and Bell P. R. (1970) Primordial radionuclide abundances, solar proton and cosmic-ray effects, and ages of Apollo lunar samples by nondestructive gamma ray spectrometry. *Proc. Apollo 11 Lunar Sci. Conf.*, *Geochim. Cosmochim. Acta*, Suppl. 1, Vol. 2, pp. 1407–1423. Pergamon.
- O'Kelley G. D., Eldridge J. S., Schonfeld E., and Bell P. R. (1971) Cosmogenic radionuclide concentrations and exposure ages of lunar samples from Apollo 12. *Proc. Second Lunar Sci. Conf.*, *Geochim. Cosmochim. Acta*, Suppl. 2, Vol. 2, pp. 1747–1755. MIT Press.
- Perkins R. W., Rancitelli L. A., Cooper J. A., Kaye J. H., and Wogman N. A. (1970) Cosmogenic and primordial radionuclide measurements in Apollo 11 lunar samples by nondestructive analysis. *Proc. Apollo 11 Lunar Sci. Conf.*, *Geochim. Cosmochim. Acta*, Suppl. 1, Vol. 2, pp. 1455–1469. Pergamon.

- Rancitelli L. A., Perkins R. W., Felix W. D., and Wogman N. A. (1971) Erosion and mixing of the lunar surface from cosmogenic and primordial radionuclide measurements in Apollo 12 lunar samples. *Proc. Second Lunar Sci. Conf.*, *Geochim. Cosmochim. Acta*, Suppl. 2, Vol. 2, pp. 1757–1772. MIT Press.
- Rancitelli L. A., Perkins R. W., Felix W. D., and Wogman N. A. (1972) Lunar surface processes and cosmic ray characterization from Apollo 12-15 lunar sample analyses. *Proc. Third Lunar Sci. Conf.*, *Geochim. Cosmochim. Acta*, Suppl. 3, Vol. 2, pp. 1681-1691. MIT Press.
- Shedlovsky J. P., Honda M., Reedy R. C., Evans J. C., Jr., Lal D., Lindstrom R. M., Delany A. C., Arnold J. R., Loosli H., Fruchter J. S., and Finkel R. C. (1970) Pattern of bombardment-produced radionuclides in rock 10017 and in lunar soil. *Proc. Apollo 11 Lunar Sci. Conf.*, *Geochim. Cosmochim. Acta*, Suppl. 1, Vol. 2, pp. 1503–1532. Pergamon.